Nanotechnology with Atom Optics

Jabez J. McClelland, Shannon B. Hill, Marin Pichler, and Robert J. Celotta

Electron Physics Group

National Institute of Standards and Technology

Gaithersburg, MD 20899-8412 USA

Abstract

A brief review of atom optics is presented, with emphasis on how it can be applied in the field of nanotechnology. Two specific examples are discussed: laser-focused atomic deposition and deterministic production of single atoms. Results are summarized for these two techniques, and discussion is presented of how they can impact progress in the development of nanotechnology.

Keywords: Atom optics; laser-focused atomic deposition; atom lithography; atoms on demand; single atoms; chromium

I. Introduction

Since its beginnings roughly two decades ago, nanotechnology has evolved from a rather narrowly defined area, concerned primarily with microelectronics, into an immense field that interfaces with areas ranging from physics and chemistry to materials science and biology. Throughout the growth of the field, a central theme that has emerged is the need to develop new ways to control and manipulate matter on scales that are measured in nanometers. That is, there is a need for methods that can be used to reliably and reproducibly build structures consisting of just a few to several hundred atoms. To meet this need, a number of new techniques have been developed which approach the problem of nanofabrication from directions that differ significantly from the conventional approach of top-down lithography. These new techniques, which include, for example, molecular self-assembly and scanned-probe manipulation, have not so much simply pushed the resolution limits for fabrication (lithography itself has evolved well into the nanometer regime) as they have opened a wide range of new possibilities for research and technological development on the nanoscale. The result has been a surprising diversification of nanotechnology, and a realization that the more varied the toolbox of techniques for nanofabrication is, the more opportunities there are for new, unforeseen developments.

A unique approach to nanofabrication that has seen development over the past decade or so is the use of atom optical techniques to build nanostructures. The notion of atom optics refers to the manipulation of beams of neutral atoms in ways analogous to the ways that beams of light are manipulated in conventional optics – with lenses, mirrors,

beamsplitters, etc. The idea is to use these manipulation techniques to control the motion of neutral atoms with nanoscale precision to implement novel nanofabrication schemes.

Using neutral atoms for nanofabrication has a number of distinct advantages over other methods, such as photon, electron, or ion beam lithography. Because neutral atoms are massive compared with photons or electrons, they tend to have a very small de Broglie wavelength, and hence resolution limits are not affected by diffraction effects. Because they are charge-neutral, unlike electrons or ions, neutral atoms are not affected by the space charge effects that can make it very difficult to concentrate many particles into a very small region. These points suggest that the resolution of atom-optical nanofabrication could, at least in principle, be better than conventional methods. However, perhaps the most intriguing advantage of atom optical methods is the fact that it opens the possibility of building nanostructures directly in a one-step process by assembling atoms, instead of carving them out of bulk material in a multistep process, as is done in lithography. The result is a fabrication method that is potentially more efficient, significantly reduces the possibility of contamination with resist or ion beam materials, and may lead to a range of new capabilities.

The treatment of neutral atom manipulation as a form of optics has emerged from the extensive body of research in laser cooling and trapping of neutral atoms that has been under development since the early 1980s [1]. While it may at first seem counterintuitive that a strong enough force can be exerted on a neutral atom to significantly affect its trajectory, it turns out that if a laser is tuned near a resonance absorption line of the atom, the interaction between the electromagnetic field and the atom can in fact have a considerable effect on the motion. The source of this effect is

momentum transfer from the light field to the atom, which occurs in two ways: (1) the atom can absorb photons with specific momentum from the laser, and reradiate them into all directions, resulting in a net gain of momentum for the atom; and (2) the oscillating electric field of the light can induce an oscillating electric dipole moment on the atom, which then generates a force in the presence of any gradient in the electric field that might be present.

The first of these momentum transfer mechanisms, often referred to as the spontaneous force, is a stochastic force, in the sense that there is a net force only on average, and there is an accompanying diffusion of the atom's momentum. When implemented with a laser that is tuned slightly below an atomic resonance, this force combines with the Doppler shift, which causes atoms with larger velocities traveling toward the light source to feel stronger forces, to result in the phenomenon known as laser cooling. Laser cooling can have dramatic effects on beams of atoms, collimating them to sub-milliradian divergences, or slowing and confining them in traps where their temperature can be reduced to the order of $100~\mu K$ or even less. The result is a level of control over neutral atom motion that can open a wide range of possibilities for nanotechnology.

In contrast with the spontaneous force, the second momentum transfer mechanism, which is commonly referred to as the dipole force, is conservative and is often written in terms of a potential energy. Since it relies on resonant excitation of an atomic dipole moment, the sign and magnitude of the dipole force depends on the detuning of the laser from the atomic resonance. If the laser frequency is detuned below the atomic resonance, the induced dipole oscillates in phase with the electric field, and

the force is felt in a direction toward higher electric field strength (or, equivalently, toward higher laser intensity). If the laser is detuned above resonance, the opposite holds true and the force is towards lower intensity. Provided the laser is not too intense or tuned too closely to resonance, the dipole potential can be written as

$$U = \frac{\hbar \Gamma^2}{8\Delta} \frac{I}{I_s} \tag{1}$$

where Γ is the atomic transition probability, Δ is the detuning of the laser from resonance, I is the laser intensity, and I_s is the atomic saturation intensity, given by $\pi h c \Gamma/(3\lambda^3)$ (h is Planck's constant, c is the speed of light, and λ is the wavelength of the light). Equation (1), which is valid for $(I/I_s)[\Gamma^2/(\Gamma^2+4\Delta^2)]<<1$, shows a linear dependence of the potential on laser intensity, and an inverse proportionality to the detuning. These simple dependencies provide a straightforward conceptual basis for constructing atom optical focusing elements with laser beams: the intensity distribution must mimic the desired potential energy surface, while the total intensity and detuning provide a potential strong enough to bend the atomic trajectories in the desired way.

The use of atom optics for nanotechnology has been a subject of research by a number of groups for about a decade, and several review articles have been published which contain a great level of detail [2,3]. In addition, review articles covering more general aspects of atoms optics such as reference [4] are available. For the purposes of the present paper, we will discuss two applications that have been investigated in our laboratory, as examples of the possibilities that atom optics can present for nanotechnology.

II. Laser-focused atomic deposition

Laser focused atomic deposition is a process that makes use of both laser cooling and the dipole force to deposit nanostructures directly onto a surface. In essence, a beam of atoms is collimated by a region of laser cooling and then passes through a laser standing wave detuned above resonance propagating across the surface (see Fig. 1). In the standing wave, the atoms experience a dipole potential that exerts a force toward the low intensity regions, or nodes, of the standing wave. Because the intensity, and hence the potential energy, depends approximately quadratically on transverse position near a node, the dipole force is a restoring force with magnitude linearly proportional to the transverse distance from the node. This is the condition required for Gaussian focusing, and the result is that each node of the standing wave acts as a nanoscale lens for atoms. Since the lens itself has a size equal to half the laser wavelength, the focal spot can be much smaller. In fact, simulations show [5] that with a perfectly collimated, monoenergetic beam of atoms, spots as small as a few nanometers can be obtained. Of course in reality, the atomic beam will have some residual divergence and will in general contain atoms with a range of kinetic energies. The focusing process then becomes a little more complex, with multiple focusing (or "channelling") occurring to a varying degree, depending on laser intensity, detuning, and spatial profile.

The fabrication of permanent nanostructures by laser-focused atomic deposition was first demonstrated using chromium atoms with laser light provided by a single-frequency, stabilized dye laser operating with stilbene-3 laser dye and tuned near the 7S_3 – $^7P_4^{\,0}$ transition at 425.6 nm [6]. In this work, a series of lines with width 65 nm, height 34 nm, and spacing 212.8 nm was fabricated, covering an area of 0.4 mm × 1 mm. In

subsequent work, extension to a two dimensional array of dots was demonstrated [7,8], and a number of further extensions and improvements were carried out, such as reducing the line spacing to 53 nm [9], performing reactive ion etching to transfer the pattern to a substrate [10], replicating the pattern with polymer molding [11], and using the pattern as a template for making magnetic nanowires [12].

Two examples of chromium structures created by laser-focused atomic deposition are shown in Figs. 2a and 2b, where atomic force microscope images of patterns created by one-dimensional and two-dimensional focusing are shown, respectively. The lines in the one-dimensional array represent some of the narrowest features obtained with chromium, with full-width at half maximum of (29 ± 3) nm. Based on extensive measurements and modeling of the laser focusing process, there is strong evidence that this width is limited by the growth properties of chromium, rather than the atom optical resolution [13].

The process of laser-focused deposition continues to see new developments and applications as research progresses. One clear application is to use the periodic arrays of deposited lines as nanoscale pitch standards, since the periodicity can be traced to a very well-determined optical frequency [14,15]. Periodic doping of materials by codeposition during growth also appears to hold a great deal of promise [16]. The extension to laser focusing of other material besides chromium has already been demonstrated with aluminum [17] and ytterbium [18], and work is progressing on gallium and indium [19], as well as iron [20]. Another obvious generalization of these techniques would be the generation of truly arbitrary patterns, as opposed to periodic patterns. To date, this

remains a challenge, though some degree of flexibility appears to be possible using more complicated superpositions of laser beams [21].

III. Determinsitic source of single atoms

As another example of how atom optics can be put to use to introduce new techniques into nanotechnology, we discuss the recent demonstration of a deterministic source of single atoms in our laboratory [22]. Absolute control over single atoms might be considered the ultimate goal of nanotechnology, since as the sizes of structures continue to shrink in the nano regime, it will eventually be important to control the exact placement of every atom. To exercise absolute control over atoms, not only is it important to have atomic scale resolution, but it is also necessary to have control of the precise number of atoms and the times at which they arrive. For this to be possible, a source of "atoms on demand" must be developed, so that single atoms can be produced deterministically when they are needed.

Our atom-on-demand source is based on a magneto-optical trap (MOT) for chromium atoms [23]. A MOT makes use of the spontaneous force exerted by three mutually orthogonal pairs of circularly-polarized laser beams to capture and cool a population of atoms. The laser beams, tuned near to, but just below, an atomic resonance, are aimed at the zero point in the center of a region of magnetic field that increases in magnitude in all directions. The field is produced by a pair of coaxial coils, spaced about a radius apart, with oppositely flowing currents (anti-Helmholtz configuration). Because of the Zeeman effect and the handedness of the circular polarization in the laser beams, atoms that move out of the center region are shifted closer

to resonance with a laser beam that tends to push them back to the center. The result is a restoring force that keeps atoms trapped at the center. Because the lasers are tuned below the atomic resonance, there is also a cooling effect on the trapped atoms, and their temperature can reach a very low level, approaching $100~\mu K$ or so, depending on the atomic species being trapped.

The MOT used for our deterministic single-atom source is constructed in a vacuum system with very low scattered light and a high efficiency optical collection system for detecting the resonance fluorescence emitted by the atoms while they are in the trap. With this arrangement, the fluorescence from a single atom is easily discernable, with a count rate of about (3000 to 4000) s⁻¹ above a background level of approximately 100 s⁻¹. The principle of operation of the deterministic source is to sense the presence or absence of a single atom in the MOT, and to use this information to control the loading and loss of the MOT: if there are no atoms present, turn on loading; if there are one or more atoms, turn off loading; if there are two or more atoms, dump the MOT and reload as quickly as possible. The result is a feedback-controlled population in the MOT that can be held at the single-atom level with very high probability, essentially indefinitely.

Loading of the magneto-optical trap is accomplished by evaporating chromium atoms from a high-temperature effusion cell, which generates a thermal beam of atoms. In order to be able to rapidly control loading into the MOT, an optical pumping scheme is used that places all atoms that would ordinarily be loaded from the beam into a metastable state. In this state the atoms are no longer resonant with the MOT laser beams, so they will not be trapped. When loading is desired, another laser, intersecting

the atom beam just before the MOT, can be turned on to optically pump the atoms back to the ground state, whence they can be trapped. Dumping of the trap when the population goes above the single atom level is achieved by either momentarily shifting the MOT laser frequency above resonance, or by blocking one of the MOT beams and momentarily shutting off the magnetic field.

Operation of the deterministic single-atom source is illustrated in Fig. 3, where time series and histograms of the fluorescence detected from the MOT are shown both with and without the feedback mechanism active. With no feedback, the population of atoms follows a random walk, as atoms enter and leave the trap based on Poisson-distributed load and loss processes (loss in this case occurs predominantly because residual gas molecules in the vacuum chamber collide with the trapped atoms). With feedback turned on, the trap population remains locked at the single-atom level, with only occasional fast excursions to the zero-atom or two-atom levels. Averaged over a 200 s time interval, the highest fraction of time spent in the single-atom state observed for our experimental conditions was $(98.7 \pm 0.1)\%$, as shown in Fig. 3.

Measurements were also carried out examining the probability of ejecting a single atom when the trap contents were dumped periodically, as a function of the rate of dumping. We found that the probability dropped slowly with increasing rate, as expected, but maintained a minimum value of 90% at rates as high as 10 Hz. We also performed Monte-Carlo simulations of the entire feedback-controlled MOT system, and found excellent agreement with our measurements. These simulations showed that with further optimization of the system, a single-atom probability of 99% could be maintained at extraction rates of up to 400 Hz.

IV. Conclusions

The two techniques described in this paper, laser-focused atomic deposition and deterministic production of single atoms, represent two illustrations of the diverse ways in which atom optics can be put to use for nanotechnology. Other possibilities exist; for example, there is a large body of literature on atom lithography using alkali atoms or metastable rare gas atoms (see reference [3] and references therein), and there are some very interesting developments in atom holography [24] that may have a significant impact. Also, atom optics is a rapidly advancing field, and there are a number of new developments such as Bose-Einstein condensation [25] and trapping of atoms in microscopic magnetic traps [26] that could find new applications in nanotechnology as research continues. As these and other methods for manipulating neutral atoms emerge, it appears clear that the ever-widening field of nanotechnology will continue to benefit from the infusion of new approaches stemming from fundamental research in atom optics and related neutral atom manipulation techniques.

- 11 -

References

- [1] See, e.g., H. J. Metcalf and P. van der Straten, <u>Laser Cooling and Trapping</u> (Springer, New York, 1999).
- [2] J. J. McClelland, in Handbook of Nanostructured Materials and Nanotechnology,
- v. I, ed. by H. S. Nalwa (Academic Press, San Diego, CA, 2000), p. 335-385.
- [3] D. Meschede and H. Metcalf, J. Phys. D 36, R17 (2003).
- [4] C. S. Adams, M. Sigel, and J. Mlynek, Phys. Rep. 240, 143 (1994).
- [5] J. J. McClelland, J. Opt. Soc. Am. B 12, 1761 (1995).
- [6] J. J. McClelland, R. E. Scholten, E. C. Palm, and R. J. Celotta, Science 262, 877 (1993).
- [7] R. Gupta, J. J. McClelland, Z. J. Jabbour, and R. J. Celotta, Appl. Phys. Lett. **67**, 1378 (1995).
- [8] U. Drodofsky, J. Stuhler, Th. Schulze, M. Drewsen, B. Brezger, T. Pfau, and J. Mlynek, Appl. Phys. B **65**, 755 (1997).
- [9] R. Gupta, J. J. McClelland, P. Marte, and R. J. Celotta, Phys. Rev. Lett. **76**, 4689 (1996).
- [10] J. J. McClelland, R. Gupta, R. J. Celotta, and G. A. Porkolab, Appl. Phys. B 66, 95 (1998).

- [11] Y. Xia, J. J. McClelland, R. Gupta, D. Qin, X.-M. Zhao, L. L. Sohn, R. J. Celotta, and G. M. Whitesides, Adv. Mater. 9, 147 (1997).
- [12] D. A. Tulchinsky, M. H. Kelley, J. J. McClelland, R. Gupta, and R. J. Celotta, J. Vac. Sci. & Tech. A 16, 1817 (1998).
- [13] W. R. Anderson, C. C. Bradley, J. J. McClelland, and R. J. Celotta, Phys. Rev. A 59, 2476 (1999).
- [14] J. J. McClelland, W. R. Anderson, C. C. Bradley, M. Walkiewicz, R. D. Deslattes,E. Jurdik, and R. J. Celotta, J. Res. Natl. Inst. Stand. Tech. 108, 99 (2003).
- [15] E. Jurdik, H. van Kempen, J. Hohlfeld, Th. Rasing, and J. J. McClelland, Appl. Phys. Lett. **80**, 4443 (2002).
- [16] Th. Schultze, T. Müther, D. Jürgens, B. Brezger, M. K. Oberthaler, T. Pfau, and J. Mlynek, Appl. Phys. Lett. 78, 1781 (2001).
- [17] R. W. McGowan, D. M. Giltner, and S. A. Lee, Opt. Lett. 20, 2535 (1995).
- [18] R. Ohmukai, S. Urabe, and M. Watanabe, Appl. Phys. B 77,415 (2003).
- [19] S. J. Rehse, R. W. McGowan, and S. A. Lee, Appl. Phys. B **70**, 657 (2000).
- [20] E. T. Sligte, B. Smeets, R. C. M. Bosch, K. M. R. van der Stam, L. P. Maguire, R. E. Scholten, H. C. W. Beijerinck, and K. A. H. van Leeuwen, Microelectronic Engineering 67, 664 (2003).
- [21] M. Mützel, U. Rasbach, D. Meschede, C. Burstedde, J. Braun, A. Kunoth, K. Peithmann, and K. Buse, Appl. Phys. B 77, 1 (2003).
- [22] S. B. Hill and J. J. McClelland, Appl. Phys. Lett. **82**, 3128 (2003).
- [23] C. C. Bradley, J. J. McClelland, W. R. Anderson, and R. J. Celotta, Phys. Rev. A **61**, 053407 (1999).

[24] F. Shimizu, Adv. At. Mol. Opt. Phys. 42, 73, 2000.

[25] J. R. Anglin and W. Ketterle, Nature **416**, 211 (2002).

[26] E. A. Hinds and I. G. Hughes, J. Phys. D 32, R119 (1999).

Figure Captions

Figure 1. Schematic of laser-focused atomic deposition. An effusive thermal source produces a mechanically collimated beam of atoms that is further collimated by transverse laser cooling. The atoms then pass through a near-resonant laser standing-wave propagating parallel to and just above a substrate surface. The atoms are focused by the nodes of the standing wave into an array of nanoscale lines as they deposit onto the surface. The laser-cooling beam and the standing wave laser beam are typically produced by the same laser, with a frequency shift provided by an acousto-optic modulator (AOM).

Figure 2. Atomic force microscope images of nanostructures created by laser-focused atomic deposition. (a) one-dimensional pattern, showing 29 nm feature width. (b) two dimensional pattern, created by crossing two one-dimensional standing waves at 90°. Note the vertical scale of the images is not the same as the horizontal scales.

Figure 3. Performance of a deterministic single-atom source based on a feedback-controlled magneto-optical trap. (a) Time series of the fluorescence signal from the trap with feedback turned off, showing clear steps corresponding to zero, one, two, three, and four atoms, and a random walk in trap population. (b) Time series of the fluorescence signal from the trap with feedback turned on, showing nearly constant fluorescence at the single-atom level. (c) Histogram of feedback-off data shown in (a). (d) Histogram of feedback-on data shown in (b), showing a 98.7% single-atom occupancy.

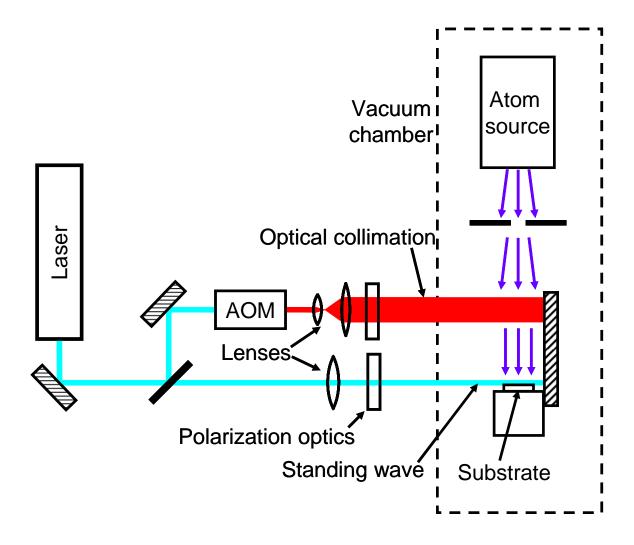
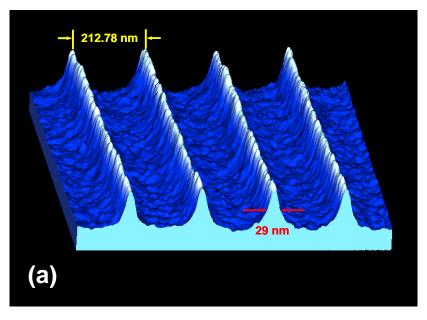


Figure 1. Schematic of laser-focused atomic deposition. An effusive thermal source produces a mechanically collimated beam of atoms that is further collimated by transverse laser cooling. The atoms then pass through a near-resonant laser standing-wave propagating parallel to and just above a substrate surface. The atoms are focused by the nodes of the standing wave into an array of nanoscale lines as they deposit onto the surface. The laser-cooling beam and the standing wave laser beam are typically produced by the same laser, with a frequency shift provided by an acousto-optic modulator (AOM).



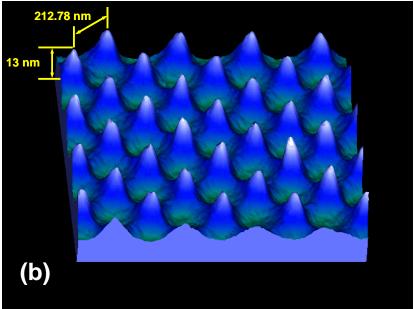


Figure 2. Atomic force microscope images of nanostructures created by laser-focused atomic deposition. (a) one-dimensional pattern, showing 29 nm feature width. (b) two dimensional pattern, created by crossing two one-dimensional standing waves at 90°. Note the vertical scale of the images is not the same as the horizontal scales.

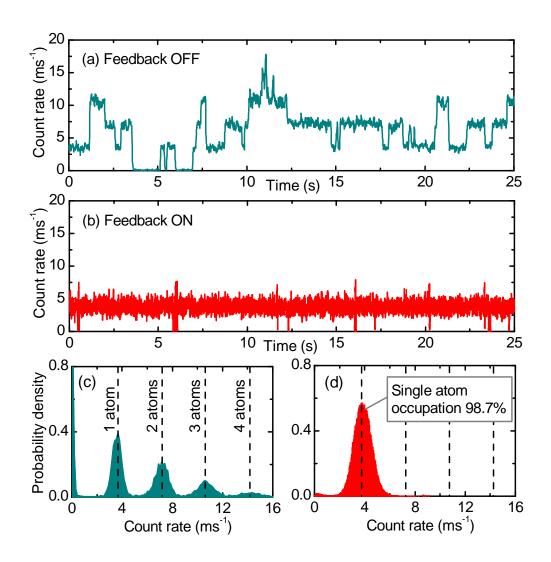


Figure 3. Performance of a deterministic single-atom source based on a feedback-controlled magneto-optical trap. (a) Time series of the fluorescence signal from the trap with feedback turned off, showing clear steps corresponding to zero, one, two, three, and four atoms, and a random walk in trap population. (b) Time series of the fluorescence signal from the trap with feedback turned on, showing nearly constant fluorescence at the single-atom level. (c) Histogram of feedback-off data shown in (a). (d) Histogram of feedback-on data shown in (b), showing a 98.7% single-atom occupancy.